DOUBLE BETA DECAY OF U-238

PROGRESS REPORT
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Abstract

During the last period, two samples of 33 yr old uranyl nitrate have been examined for the accumulation of Pu-238 formed as a result of double beta decay. The 2.5 kg sample has led only to an upper limit of 6×10^{20} yr. The 8.5 kg sample has produced positive results corresponding to a half-life of 2×10^{21} yr. The possibility of contaminating nuclear reactions has been carefully examined and eliminated. The possibility that contamination may have been introduced by the large quantities of chemicals that were used is being investigated.

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Introduction

This report covers the period from 1988 December 29 (the date of the last report on this project) to 1990 November 29. It includes sections on (a) continuing attempts to find and maintain stable alpha particle detector systems of high efficiency but low background; (b) data analysis of an experiment to determine the cross-section of the $^{238}\text{U} (p,n) ^{238}\text{Np}$ reaction; (c) evaluation of possible nuclear reactions leading to a false signal of the double beta reaction; and (d) the results of two experiments on 33 yr old uranium, the last one providing a positive result for the ^{238}U double beta decay half-life.

Low-Level Alpha Counters

This program has made use of alpha detection systems at Los Alamos and at Chicago. Although all were chosen to have low backgrounds (≤ 0.3 events/day) the long measurements involved in this program (typically many months) have revealed large differences in the efficiencies, backgrounds and the stability of the backgrounds and associated electronics.

The Los Alamos counter has a relatively high efficiency and regular supervision. This has revealed a high and variable background and some electronics instability.

One of the Chicago counters has the highest efficiency but also a high, but stable, background.

The other Chicago counting system is of relatively low efficiency but has almost 6 times lower background (~ 1 event in 70 days) in the ^{238}Pu alpha region. The detector for this system was made from silicon produced before ~ 1967 .

Because of the difficulty in guaranteeing the background and stability of the counting system over the long measurement times of this experiment, the final samples from the work were measured on all three counting set-ups.

The $^{238}\text{U} (p,n) ^{238}\text{Np}$ Cross-Section

In spite of almost 50 years of study of the nuclear properties and reactions of the heaviest nuclides, the cross-section of the $^{238}\text{U} (p,n) ^{238}\text{Np}$ reaction has not been published. Because of the importance of this reaction in evaluating the possible production of ^{238}Pu by nuclear processes other than double beta decay (see below), an experiment was performed in 1987 at Los Alamos to determine this cross-section in the energy range 6-12 MeV. Preliminary analysis of the data at the time indicated that this cross-section was low enough so that competing reactions were not important in addressing double beta decay half-lives in the 10^{20} yr range.

As the experimental limits on the double beta decay half-life reached the 10^{21} yr range, the possibility of competing reactions had to be evaluated more carefully. The data from the 1987 cross-section experiment has been worked up. They lead to cross-sections of 0.04 ± 0.01 , 1.3 ± 0.2 , 2.8 ± 0.2 , and 5.1 ± 0.5 mb at 5.4, 7.5, 9.6, and 11.6 MeV respectively. These cross-sections allow quantitative evaluation of the production of ^{238}Pu by cosmic rays, the natural radioactivity of uranium, and by external neutrons. The most serious of these contributes less than 10% to the observed double beta decay signal.

As a byproduct of these cross-section experiments, the relative yields of products of the 12 MeV proton fission of ^{238}U were determined. These relative yields of fission products match quantitatively the distribution found in the 14 MeV *neutron* fission of ^{238}U . This is consistent with the idea that the mass and excitation energy of the fissioning nuclide is the most important factor in determining the relative yields of different fission products.

Finally, the data with 6 MeV protons leads to a ratio of fission to neutron emission probabilities agreeing with those deduced for the same excitation of $^{238}\text{Np}^*$ by charged particle studies. The present work extends data on this competition to higher excitation energies.

Competing Reactions Leading to ^{238}Pu

In addition to double beta decay of ^{238}U , there are other nuclear processes that might lead to the production of the same nuclide, particularly in large samples of uranyl nitrate stored at close to sea level for 33 years.

The most serious of these is the $^{238}\text{U} (p,n) ^{238}\text{Np}$ reaction, since the ^{238}Np would then beta decay to ^{238}Pu . Using the (p,n) cross-section determined (see above), the probability of conversion of a proton to ^{238}Np in uranyl nitrate has been evaluated to increase from less than 10^{-7} at 6 MeV to 1.3×10^{-6} at 14 MeV. In spite of the high radioactivity of the large sample, neither the alpha activity nor the spontaneous fission can produce even 10^5 atoms of ^{238}Pu in 33 years.

Similarly, the contribution of cosmic rays in transforming ^{238}U to ^{238}Np has been evaluated. R. Davis⁽¹⁾ provided data on the production of ^{37}Ar from ^{37}Cl and of ^{71}Ge from ^{71}Ga in large samples by cosmic rays at sea level. These data have been converted to the present situation using the cross-section data for the respective (p,n) reactions and the stopping power of the media involved. The result is a safe upper limit for production by cosmic rays.

The possibility that radiation from the 450 MeV Chicago cyclotron could have produced ^{238}Pu has been evaluated using the radiation exposure records kept in the neighborhood of the stored uranyl nitrate sample. This provides the most uncertain result, but the upper limit is less than 10% of the observed ^{238}Pu signal.

Results of Two Experiments on Double Beta Decay of ^{238}U

The plutonium that had accumulated in 32 yr in 2.5 kg of uranyl nitrate (Expt. A-4) was isolated chemically using the added ^{239}Pu tracer to measure the efficiency of recovery and measurement. Essentially the same chemistry was used as in the previous work using 600 gm of the same material.⁽²⁾ The final sample was of poorer quality than that obtained in the previous experiment.

Extensive alpha measurements (more than 200 days) showed no evidence for the presence of the 5.5 MeV alpha particles of ^{238}Pu . The result was a 90% confidence upper limit of 6×10^{20} yr for the double beta decay half-life of ^{238}U . This was a factor of 10 longer than that reported from the 600 gm sample.⁽²⁾

This result was presented by M. Moe⁽³⁾ as part of his invited summary talk at the June 1990 Neutrino conference in Geneva, Switzerland.

The plutonium from the 8.5 kg sample (A-5) was isolated using more involved chemistry than previously used. This was done in an attempt to improve the yield from the larger initial sample, and to provide a higher quality final sample.

The measurements on all three alpha detecting systems on this sample agree in giving a positive signal of 0.09 events per day of ^{238}Pu in the sample. This corresponds to the production of 2×10^5 atoms of plutonium from 10^{25} atoms of U in 33 years. The measurements on this sample have survived examination for possible contaminating radioactivities. They correspond to a half-life for double beta decay of ^{238}U of 2×10^{21} yr. This work has been written up and submitted for publication to *Physical Review Letters*.

Because of the large sample, the associated large amounts of chemicals and the extra chemical steps in this experiment, these should be examined for possible introduction of fall-out ^{238}Pu or other contamination. This will be done.

The value of 2×10^{21} yr is longer than previous theoretical estimates. However, within the last year Klapdor and coworkers⁽⁴⁾ have published new estimates that predict a value of only $\sim 10^{23}$ yr for the 2ν double beta decay of this nuclide. Their estimate for a 0ν mode would imply that the reabsorbed neutron would have a mass of ~ 12 eV to explain the present experimental results. Although much larger than the 1.6 eV neutrino mass deduced⁽⁵⁾ from not seeing neutrinoless double beta decay of ^{76}Ge , the present experiment involves different matrix elements and addresses the possibility of other modes of double beta decay.

References

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